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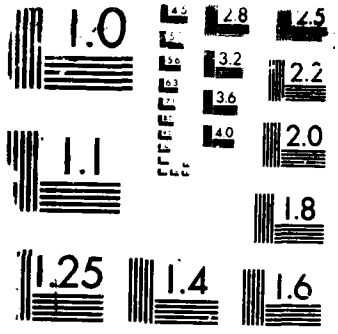

MICROCOPY RESOLUTION TEST CHART

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DISCHARGE EXCITATION OF DYE VAPORS

FINAL REPORT

by

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September 1987

Prepared for

U. S. Army Research Office
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Experiments have been conducted to assess the feasibility of obtaining a discharge-pumped dye vapor laser. A 15 (or 50) cm active length, UV-preionized device has been developed which exhibits a specific power loading of the medium of > 5 MW - cm^{-3} and will operate continuously at temperatures exceeding 400°C. Recently, hydrogen thyratron switching of the device and corona preionization have been installed to minimize jitter and dye fragmentation. Optimization of the rare gas/N_2 diluent mixture has been completed and fragmentation studies for several dye molecules have been conducted. POPOP and α-NPO are excellent in the latter regard but Coumarin 6 rapidly decomposes in the discharge environment. The fluorescence efficiency of α-NPO is only 40% of that for POPOP under comparable conditions. BBO and PBBO are similar in structure and molecular weight to POPOP and appear to be excellent candidates for discharge excitation. Fluorescence and small signal gain measurements are in progress.

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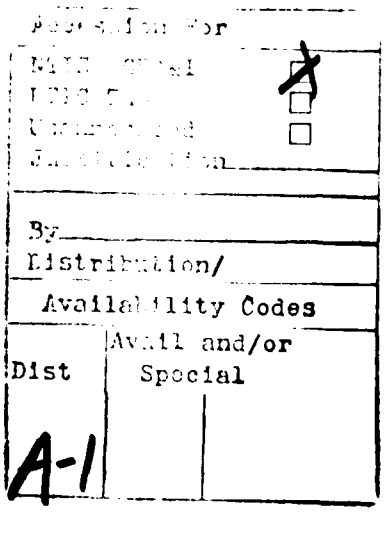
ABSTRACT

Experiments have been conducted to assess the feasibility of obtaining a discharge-pumped dye vapor laser. A 15 (or 50) cm active length, UV-preionized device has been developed which exhibits a specific power loading of the medium of ~ 5 MW - cm^{-3} and will operate continuously at temperatures exceeding 400°C. Recently, hydrogen thyratron switching of the device and corona preionization have been installed to minimize jitter and dye fragmentation. Optimization of the rare gas/N_2 diluent mixture has been completed and fragmentation studies for several dye molecules have been conducted. POPOP and α-NPO are excellent in the latter regard but Coumarin 6 rapidly decomposes in the discharge environment. The fluorescence efficiency of α-NPO is only 40% of that for POPOP under comparable conditions. BBO and PBBO are similar in structure and molecular weight to POPOP and appear to be excellent candidates for discharge excitation. Fluorescence and small signal gain measurements are in progress.



I. INTRODUCTION

The goal of this one year research program, which was sponsored by the U.S. Army MICOM through the Army Research Office, has been to assess the feasibility of obtaining a discharge-pumped, vapor phase dye laser. Considerable progress has been made towards that goal and in the following three areas in particular:

1) Improving the performance of the discharge device;

2) Optimizing the diluent mixture ($He/Ne/N_2$) for internal pumping of POPOP dye vapor; and

3) Identifying dyes other than POPOP that have POPOP's temperature and discharge stability but would lase in other spectral regions in the near-UV and visible.

Each of these areas will be described in more detail in the next section. The experimental results obtained to date are gratifying and suggest that detailed given measurements with the dyes identified under #3 above ought to be conducted in the coming fiscal year.

II. EXPERIMENTAL RESULTS

A. Discharge Device Modifications

The discharge system that was developed for these experiments is a transverse, UV-preionized device, having an active length of ~15 or 50 cm. As the experiments have progressed, several key modifications to the system have been made which have considerably improved its performance. Hydrogen thyratron switching of the discharge was recently installed that improves the jitter of the discharge by over an order of magnitude. This change was essential for being able to carry out spectroscopic and small signal gain measurements on the dye vapor plasma. Also, improvements to the oven design have reduced the time required to equilibrate the dye vapor/buffer gas mixture by a factor of two. Also, an in-situ technique for cleaning the discharge cell has been developed which is a tremendous time saver. Previously, it was necessary to remove the cell from the vacuum system, clean it with organic solvents and then heat the cell under vacuum to remove the solvent vapors. This was a time-consuming procedure but the new technique allows us to move to a new set of experiments in just a few hours.

Much of our recent work has centered on the design of the discharge's preionizer. Several experimental results have suggested that the spark array preionizer used to this point dissociates an unacceptable fraction of the dye molecules itself (prior to the main discharge). Consequently, we have conducted several tests with a corona preionizer and expect to be using it exclusively in the next few months.

B. Optimization of Diluent Mixture

Of the dyes studied to date, POPOP is (by far) the most stable at high temperature (~300°C) and exhibits the best fluorescence quantum efficiency. For this reason, we have devoted considerable effort towards optimizing the diluent gas mixture for internal pumping of POPOP dye vapor. That is, it is our intent to transfer the energy deposited in the discharge to N_2 rather than directly to the POPOP vapor. This minimizes electron impact fragmentation of the organic dye but allows the molecule to be optically pumped by spontaneous emission on the C → B band of N_2 (λ = 357.7, 380.5 nm).

Figures 1 and 2 show the results of our experiments to date. The two figures illustrate the effect on POPOP fluorescence of changing the rare gas diluent from helium to neon. Note that for He/N_2 diluent, the background fluorescence (obvious below ~230°C, the melting point for POPOP crystals) is relatively strong. In contrast, Ne/N_2-buffered mixtures yield much stronger POPOP fluorescence as evidenced by the weak background emission in Fig. 2. The latter data suggest that POPOP <u>is</u> being excited optically rather than by direct electron impact.

C. Identification of Other Dyes

A major emphasis of our recent work has been to identify dyes other than POPOP which are attractive for discharge excitation. In our last semiannual progress report, we described experiments with Coumarin 6 which clearly showed its rapid disintegration in the hostile environment of the UV-preionized discharge. Not surprisingly, therefore, dyes which behave quite nicely under

optical excitation in a liquid may be totally unsuitable for discharge pumping. Experiments conducted in the last month with BBQ in our discharge show this clearly. BBQ has a molecular weight ~2 times that of POPOP and the dye is rapidly decomposed by the discharge. However, after examining the chemical structure of more than a dozen dyes, we have identified several (such as BBO and PTP) that are very attractive for discharge excitation. All are "chain" organics having molecular weights comparable to or less than that of POPOP. It should be mentioned that all are also known to be efficient emitters when optically pumped in the gas phase.

One example that we have explored recently is α - NPO, which has a molecular weight of 271 (compared to 364 for POPOP) and a melting temperature of 104°C. Fragmentation of the vapor was observed to be low and the temperature dependence of the molecule's fluorescence intensity (λ_{max} ~396 nm) was measured (see Fig. 3). However, the maximum spontaneous emission observed from the molecule (at ~230°C) was only 40% of that obtained from POPOP. The most attractive candidates for future study are PBBO and BBO which have molecular weights of 347 and 373, respectively. Both also fluoresce primarily in the violet (λ ~400-410 nm).

In summary, then, one of the key accomplishments of this past year has been to develop a predictive capability concerning those dye molecules that are most likely to perform well in the hostile environment of the discharge. It is recommended that the identification of POPOP, BBO and PBBO as the most attractive laser candidates be followed up by intensive fluorescence and small signal gain measurements.

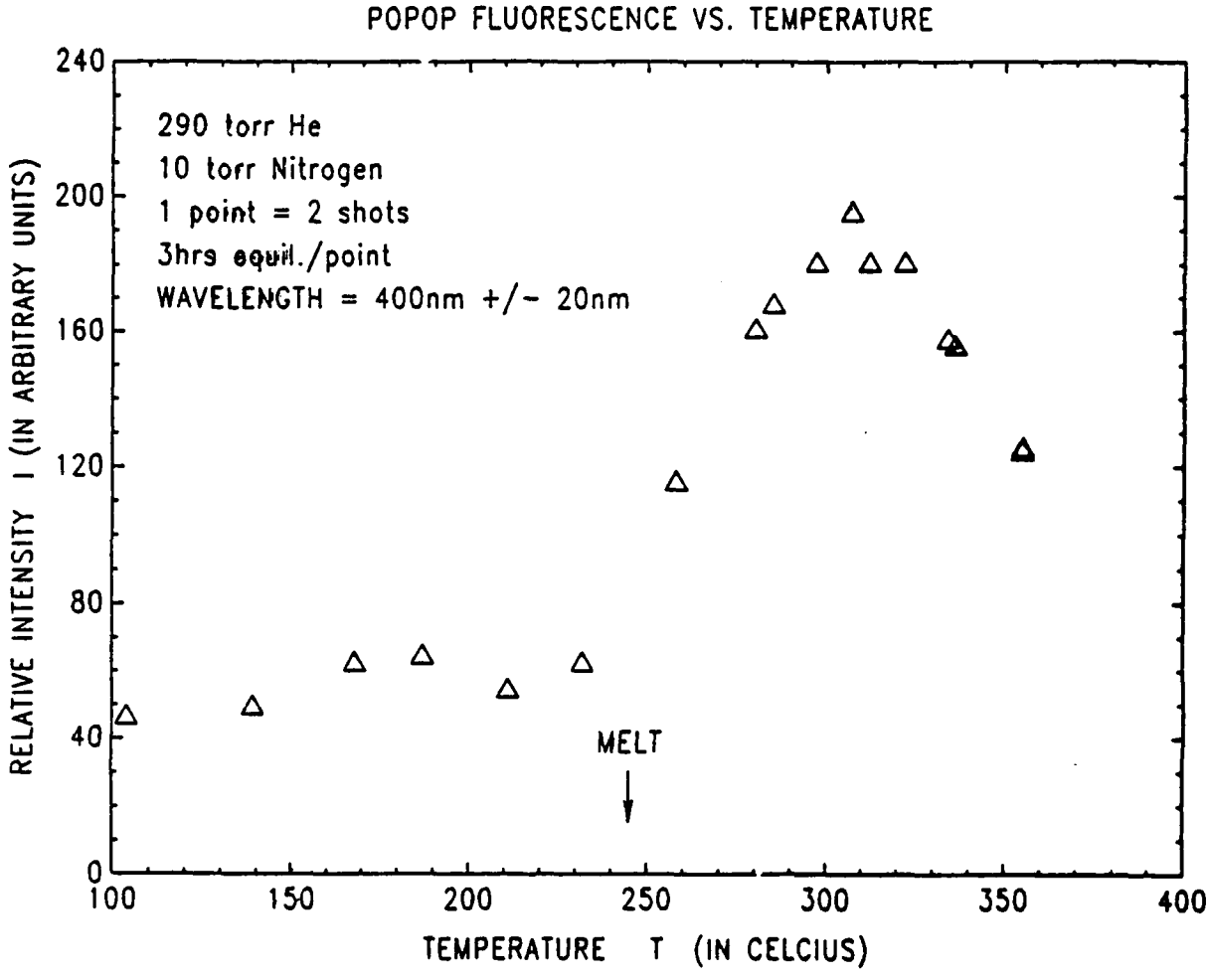


Fig. 1 Variation of POPOP violet (λ ~ 400 nm) fluorescence with the temperature of the discharge cell for the He and N_2 partial pressures (300° K) held constant at 290 and 10 Torr, respectively. Note the relatively strong background from N_2 that is evident for T $\leq$ 230° C.

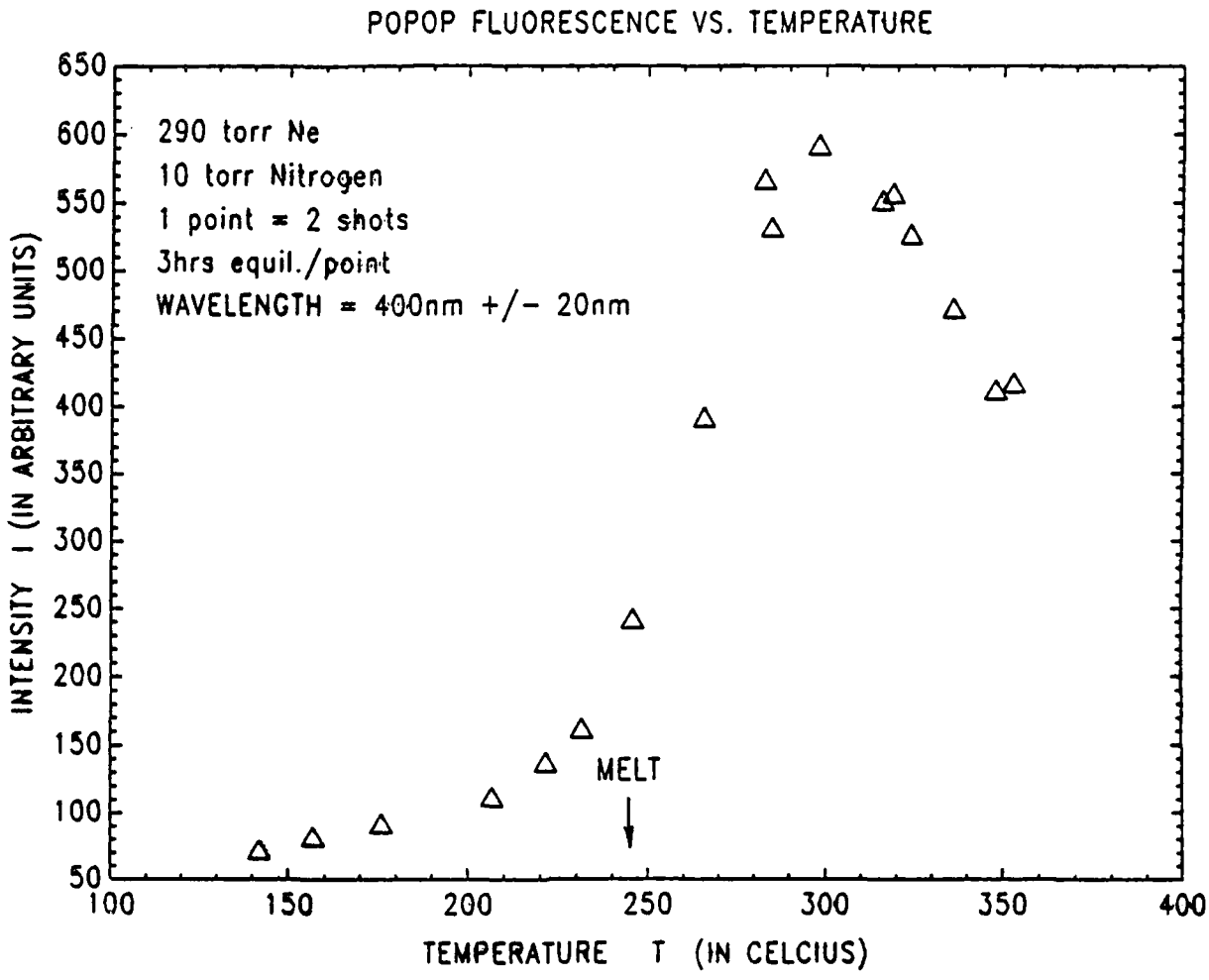
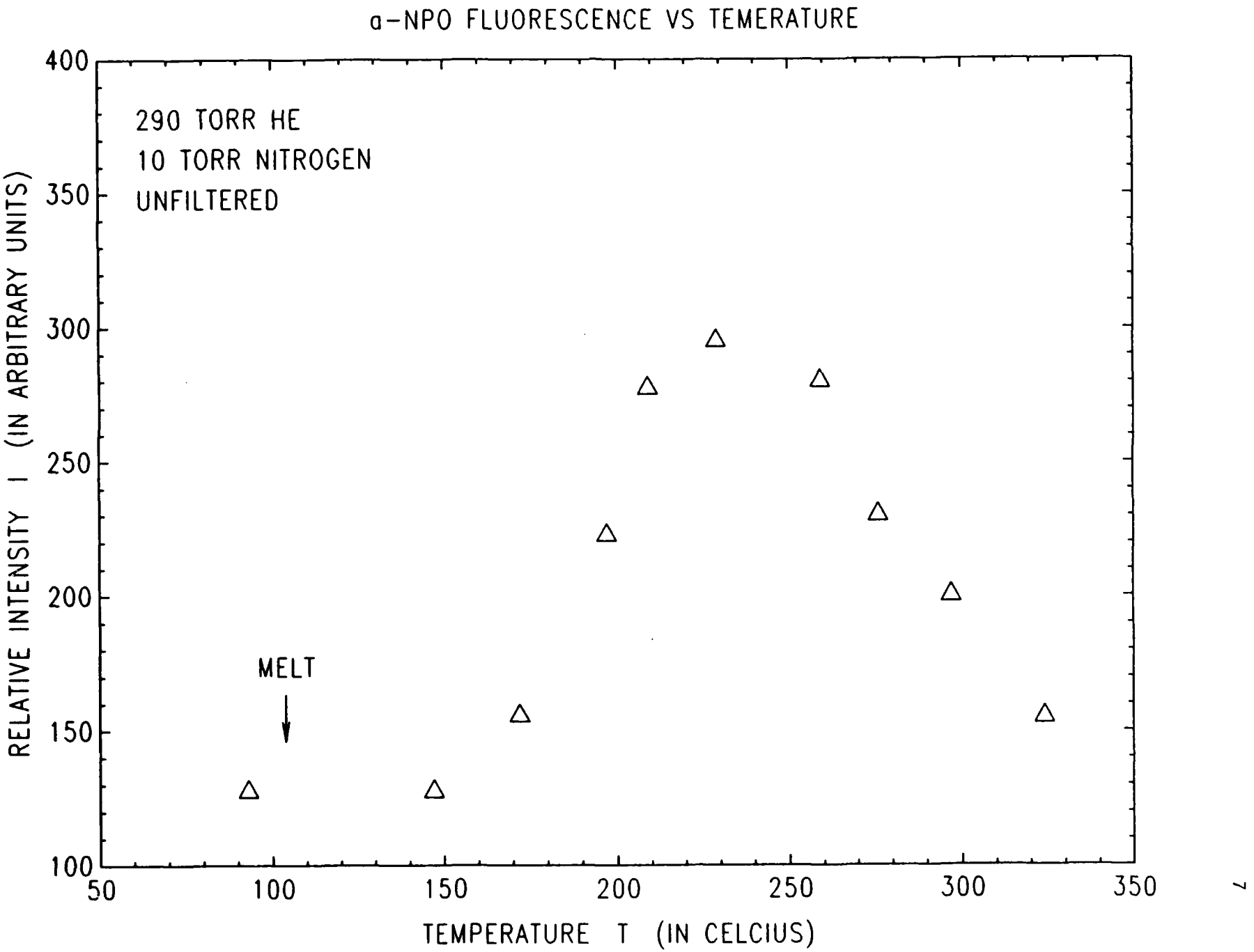


Fig. 2 Data similar to those in Fig. 1 except Ne is the rare gas diluent. The peak fluorescence intensity is roughly triple that for He. Note also the weak background fluorescence below the melting point for POPOP.


α-NPO FLUORESCENCE VS TEMERATURE
290 TORR HE
10 TORR NITROGEN
UNFILTERED
MELT
RELATIVE INTENSITY I (IN ARBITRARY UNITS)
TEMPERATURE T (IN CELCIUS)
400
350
300
250
200
150
100
50
100
150
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250
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350

Fig. 3

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